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Link to article, DOI:
10.1109/CLEOE-IQEC.2013.6801091

Publication date:
2013

Document Version
Publisher's PDF, also known as Version of record

Citation (APA):
Bache, M., Liu, X., & Zhou, B. (2013). *Few-cycle nonlinear mid-IR pulse generated with cascaded quadratic nonlinearities*. Abstract from 2013 Conference on Lasers & Electro-Optics Europe & the International Quantum Electronics Conference (CLEO/Europe-IQEC), Munich, Germany. https://doi.org/10.1109/CLEOE-IQEC.2013.6801091

Few-cycle nonlinear mid-IR pulse generated with cascaded quadratic nonlinearities

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Generating few-cycle energetic and broadband mid-IR pulses is an urgent current challenge in nonlinear optics. Cascaded second-harmonic generation (SHG) gives access to an ultrafast and octave-spanning self-defocusing nonlinearity: when $\Delta kL \gg 2\pi$ the pump experiences a Kerr-like nonlinear index change $\Delta n = n_{\rm casc} I$, where $n_{\rm casc} \propto -d_{\rm eff}^2/\Delta k$, and $d_{\rm eff}$ is the effective quadratic nonlinearity. Due to competing material nonlinearities $n_{\rm Kerr}$ the total nonlinear refractive is $n_{\rm cubic} = n_{\rm casc} + n_{\rm Kerr}$. Interestingly $n_{\rm cubic}$ can become negative (self-defocusing), elegantly avoiding self-focusing problems, and making it possible to excite solitons with normal dispersion [1].

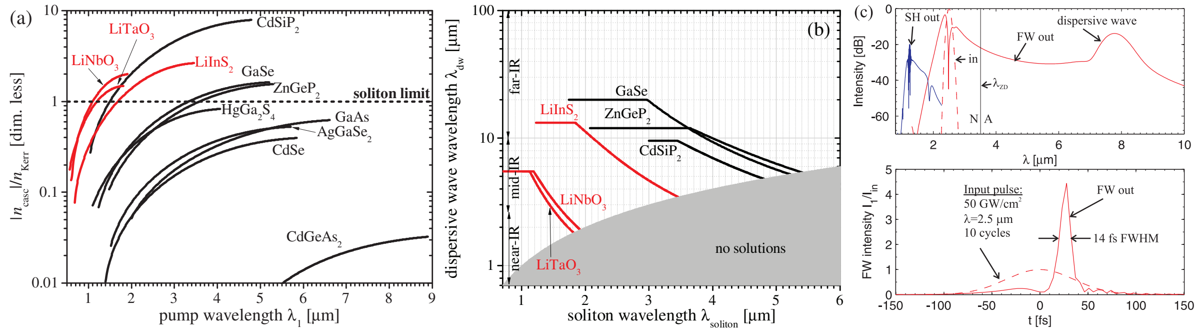

Figure 1. (a) FOM $|n_{\rm casc}|/n_{\rm Kerr}$ for type 0 cascaded SHG using dielectric (red) and semiconductor (black) materials, with $n_{\rm casc} = -2\omega_1 d_{\rm eff}^2/c^2\varepsilon_0 n^2(\omega_1)n(\omega_2)\Delta k$ and $n_{\rm Kerr}(\omega) = K' G_2(\hbar\omega/E_g)\sqrt{E_p}/n^2(\omega)E_g^4$, with the linear index $n(\omega)$, the band gap energy E_g, $K' = 7.3\times 10^{-9}$ eV$^{3.5}$m^2/W in dielectrics and $K' = 14.0\times 10^{-9}$ eV$^{3.5}$m^2/W in semiconductors, $E_p = 21$ eV is a constant. A FOM>1 is required to excite solitons. (b) The DW phase-matching curves. (c) Numerical simulation of a 10-cycle $\lambda = 2.5$ μm and $I_{\rm in} = 50$ GW/cm^2 pulse propagated 11 mm in a $LiInS_2$ crystal. 'A' and 'N' mark anomalous and normal dispersion.

Historically, *critical* (type I) cascaded SHG has been used. Recently we showed experimentally generation of strong and octave-spanning cascaded nonlinearities from a *noncritical* (type 0) interaction even without quasi-phase matching (QPM) [2]. This allows for excitation of few-cycle self-defocusing solitons at the pump wavelength, generation of octave-spanning supercontinua [2] and creation of long-wavelength Cherenkov radiation [3]. "Standard" type 0 mid-IR crystals have huge $d_{\rm eff}$, but are often overlooked because of a large Δk value which cannot be reduced (as QPM methods are not developed or applicable). This limits the strength of $n_{\rm casc}$ so it is crucial to understand whether regimes with $n_{\rm cubic} < 0$ can be found. Calculating the Kerr nonlinearity from the two-band model, Fig. 1(a) shows a figure-of-merit FOM $= |n_{\rm casc}|/n_{\rm Kerr}$; self-defocusing solitons require FOM > 1. $LiNbO_3$ and $LiTaO_3$ have an FOM>1, but at around $\lambda = 2$ μm the GVD changes sign and becomes anomalous (at this point the curves are terminated). Here the chalcogenide $LiInS_2$ and the semiconductors GaSe, $CdSiP_2$ and $ZnGeP_2$, which have large band gaps and large $d_{\rm eff}$, come into play with an FOM > 1 for $\lambda > 2$ μm. Instead for e.g. $CdGeAs_2$, its large $d_{\rm eff}$ is counteracted by a very small band gap, giving a too large $n_{\rm Kerr}$ due to the E_g^{-4} scaling. None of the crystals with FOM > 1 support self-defocusing solitons beyond $\lambda = 5.5$ μm. However, once excited the soliton will shed radiation through optical Cherenkov radiation to a linear dispersive wave (DW) in the anomalous dispersion regime $\lambda > \lambda_{\rm ZD}$. This can cover the long-wavelength range of the mid-IR. Fig. 1(b) shows the DW phase-matching curve $k_1(\omega) = k_1(\omega_{\rm sol}) - (\omega - \omega_{\rm sol})/v_{g,\rm sol}$. In Fig. 1(c) a numerical simulation of $LiInS_2$ shows that a 10-cycle input pulse at 2500 nm is soliton-compressed after 11 mm propagation to few-cycle duration. The soliton then generates a DW in the linear range (anomalous dispersion regime, $\lambda > 3.5$ μm), peaking at $\lambda = 8$ μm. This DW is very broadband and by isolating it with the equivalent of a long-pass filter we checked that it is a few-cycle pulse with excellent quality. The frequency conversion process is efficient, app. 10%. Much like what the near-IR for LN [2, 3], upon further propagation the soliton will fission into minor soliton pairs, each coupling to a dispersive wave in the anomalous dispersion regime. This will give a stronger peak around $\lambda = 8$ μm but the coherence and quality drops. Eventually a very broadband supercontinuum will form across the spectrum.

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